Direct observation of the longitudinal resonance mode in ferromagnets with random anisotropy

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Properties of a resonance mode, the existence of which is related to the amorphous structure, are reported. This mode, in addition to the uniform mode, was observed in $Co_{93-x}Zr_7(R)_x$ (R=Pr, Nd, Dy, Tb) amorphous thin films possessing an in-plane uniaxial anisotropy field H_k. A study as a function of the working frequency shows that when the magnetic field H is applied in the plane defined by the film normal and H_k, the mode can be identified as a longitudinal resonance, so detected even for $H \| h_{\rm rf} \| H_k$. The mode is localized and this localization is ''stronger'' for higher working frequencies so that the resonance fields are higher. [S0163-1829(97)01917-6]

According to theoretical computations[1–4] it should be possible to detect experimentally the longitudinal resonance (LR) in ferromagnets with random anisotropy. Indeed we reported an observation recently[5] that in a certain class of amorphous thin films two resonance modes are excited: the uniform resonance (UR) and a new resonance mode. We could verify easily that one of these mode is the uniform precession because the value of $4\pi M_s$ as computed from the resonance fields determined in perpendicular and parallel configurations agreed with static VMS measurements within some percent. However, the new resonance mode exhibited a highly unusual overall behavior. The presentation of the results was limited to the case when the steady magnetic field is applied perpendicular to the film plane ($H_\perp$). For this configuration the properties of the new mode could be interpreted by assuming that it was a longitudinal resonance (LR).

Here we report the results of new ferromagnetic resonance measurements performed at room temperature and at frequencies f=9.8, 17.9, and 35.7 GHz, respectively. These experiments allowed us to prove formally the longitudinal nature of the new mode as it is observed for the ideal theoretical configuration, i.e., when the dc field H is parallel to the rf field $h_{\rm rf}$ and also to get insights into its properties.

The LR mode is specific to ferromagnets which exhibit a random local anisotropy K_l. As opposed to a crystalline sample where the spins forming M_s are perfectly aligned along the static equilibrium direction of the magnetization $M_s^{\rm eq}$, in an amorphous material, due to the existence of K_l, the static equilibrium direction of the spins varies from site to site, so the spins are not precisely colinear. Consequently, the rotation of the spin system about any axis leads to a restoring torque. This effect can be taken into account by including a spin space rotation angle as a dynamical variable in the equations of motion. The main result of the calculation is that one finds, apart from a mode corresponding to the usual transverse configuration ($h_{\rm rf} \perp M_s^{\rm eq}$), a longitudinal one. This longitudinal mode should be excited when $h_{\rm rf}$ is parallel to $M_s^{\rm eq}$ ($h_{\rm rf} \| M_s^{\rm eq}$) and its existence is related to the slight misalignment of the spins in the equilibrium state connected with the presence of the disorder.

The new mode could be observed on amorphous $Co_{93-x}Zr_7(R)_x$ thin films where R=Nd, Pr, Dy, or Tb and $0<x<4$. The results are illustrated here by those obtained for R=Tb but the data for the other R are essentially the same. For these small R substitutions the films are ferromagnets with weak random anisotropy (FWRA),[3] and possess a rather high $4\pi M_s$ ($10^4 < 4\pi M_s < 1.2\times10^4$ Oe) and a small coercive field H_c ($0.5<H_c<3$ Oe).[6] A necessary condition for the experimental observation of the LR mode is that the films display a very well-defined in-plane uniaxial anisotropy field H_k. H_k was induced during the formation of the samples and its magnitude is $50<H_k<150$ Oe.[6] Its definition has been studied by transverse biased initial susceptibility (TBIS) measurements.[7] TBIS measurements allow one to separate the short-range intrinsic fluctuations of H_k related to K_l (the ripple), from the long-range fluctuations of H_k, the skew, which are associated with the defects. A well-resolved LR mode was observed only when the skew was negligible, a condition satisfied on a large number of samples, the features of which are reported here.

For $H_\perp$, whatever the working frequency, the UR mode is located at the high-field side of the spectra, and its main parameters, resonance field $H_\perp^{\rm UR}$ and peak-to-peak linewidth $\Delta H_\perp^{\rm UR}$, vary continously as a function of the nature and the concentration of the R: $H_\perp^{\rm UR}$ decreases and $\Delta H_\perp^{\rm UR}$ increases with increasing R contents, results in agreement with theoretical expectations.

One of the principal characteristics at 9.8 GHz of the LR is that the resonance field $H_\perp^{\rm LR}$ and linewidth $\Delta H_\perp^{\rm LR}$ were independent from the nature or the concentration of the R.[5]

The longitudinal nature of the new mode at 9.8 GHz was proved by studying its variations as a function of the angle α between H_k and $h_{\rm rf}$, the linearly polarized $h_{\rm rf}$ field being applied along the film plane. While $H_\perp^{\rm LR}$ and $\Delta H_\perp^{\rm LR}$ remain constant, the as-absorbed microwave power $P(\alpha)$ varied continuously following the law $P(\alpha)=P(0)\cos^2\alpha$, where $P(\alpha)$ is maximum for $\alpha=0$ so when $h_{\rm rf}\|H_k$. This result was explained by the fact that, as $H_\perp^{\rm LR}\ll 4\pi M_s$ ($10^3<H_\perp^{\rm LR}<1.210^3$ Oe), the magnetization vector remains very close to the film plane. Consequently $M_s^{\rm eq}$ up to first order is parallel to H_k so the condition that the LR precession should occur is satisfied because $M_s^{\rm eq}\|H_k\|h_{\rm rf}$.

The formal demonstration of the longitudinal nature of the mode requires an investigation where H is applied parallel to $h_{\rm rf}$. If $H^{\rm LR}$ is sufficiently large so that $M_s^{\rm eq}$ is oriented rigorously parallel to H, one has $H\|h_{\rm rf}\|M_s^{\rm eq}$. Moreover, this particular experimental configuration corresponds to a "nonresonant" condition: It means that the UR or any other mode related to the transverse case cannot be excited now.

The experimental setup where H can be applied parallel to $h_{\rm rf}$ was available only at 17 and 35 GHz, so the experiments were performed at these frequencies.

At 17 GHz the spectra were studied for H applied either along the film normal ($H_\perp$), so $H_\perp\perp h_{\rm rf}$, or in the film plane. For $H_\perp$ both the UR and the LR have been detected. Similarly to data obtained at 9.8 GHz, the magnitude of $P(\alpha)$ was an order of magnitude bigger for $h_{\rm rf}$ parallel to H_k, than $h_{\rm rf}$ perpendicular to H_k. For H in the film plane we studied especially the configuration $H\|H_k\|h_{\rm rf}$. The condition $M_s\|H$ was largely satisfied in accordance with the high value of the resonance field $H^{\rm LR}$ (Fig. 3), so one has $M_s\|H\|h_{\rm rf}\|H_k$. A very important result is that the values of $H^{\rm LR}$ and $\Delta H^{\rm LR}$ are the same for $H_\perp\perp h_{\rm rf}$ and $H\|h_{\rm rf}$ within experimental uncertainties. However, as shown in Fig. 1, the shape of the LR mode differs somewhat in the two configurations: It is fairly close to a Lorenztian one for $H_\perp\perp h_{\rm rf}$ and is slightly distorted for $H\|H_k\|h_{\rm rf}$. This effect is due to an unpredictable experimental difficulty. The microwave absorption by the UR is strictly equal to zero if both the external steady and the high-frequency modulation field—the use of this later one is necessary to have a sufficiently high sensitivity—is rigorously parallel to the $h_{\rm rf}$ field. Even a very small deviation with respect to this configuration, and which is very difficult to avoid, excite more or less strongly the microwave susceptibilities χ' and/or χ'' corresponding to the UR. In the parallel configuration, whatever the working frequency, fortuitously the resonance fields corresponding to the UR and to the LR are rather close and the resonance linewidth of the UR is quite large. This effect explains that the shape of the LR is perturbed slightly at 17 and more strongly at 35 GHz by the UR.

Two other experiments performed at 35 GHz allowed us

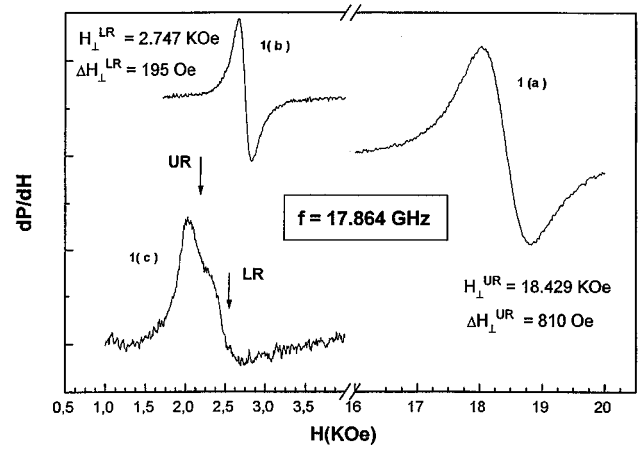

FIG. 1. Typical derivative absorption spectra at 17.864 GHz. UR (a) and LR (b) modes are for the dc field perpendicular, LR (c) is for the dc field parallel to the film plane and $H\|h_{\rm rf}$. In both configurations one has $h_{\rm rf}\|H_k$.

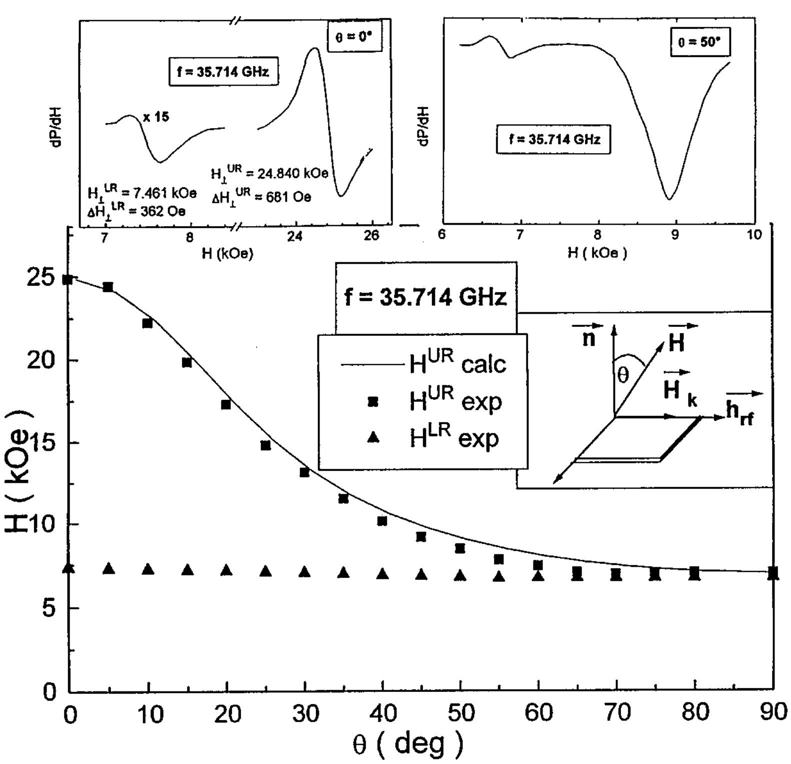

FIG. 2. Angular dependence at 35.714 GHz of the resonance fields corresponding to the UR and LR modes, respectively. The dc field is rotated in the plane formed by **n** and H_k while $h_{\rm rf}\|H_k$. The line is the computed curve corresponding to the UR. The resonance spectra for two particular values of θ are also shown.

to get more insight into the exact nature of the mode. In the first one we studied the evolution of the spectra by rotating H in the plane defined by the film normal $\vec{n}$ and H_k while h_{rf} was applied along H_k. Now the most instructive results were the variations of the respective resonance fields and that of the ratio $N = P(\text{LR})/P(\text{UR})$ of the absorbed microwave power (Fig. 2), where $P(\text{LR})$ and $P(\text{UR})$ corresponds to the LR and UR, respectively. The resonance field and linewidth related to the UR varied in agreement with the classical theoretical expectations. On the other hand, the resonance field and linewidth related to the LR remains, within experimental uncertainties, *strictly independent* from the direction of the steady field. The variations of N are also very spectacular: N increases continuously by several orders of magnitude when the parallel configuration is approached. This experiment is a direct confirmation that when the configuration $H \| h_{rf}$ is approached, $P(\text{UR})$ is continously reduced, in contrast to $P(\text{LR})$.

In the second experiment the high sensitivity of the LR for the in-plane geometry was shown. Now h_{rf} was applied parallel to H_k and the direction of H was varied in the film plane from the configuration perpendicular to $h_{rf} \| H_k$ to the configuration parallel to it. Again the results are remarkable: While the resonance field corresponding to the UR is unchanged, $P(\text{UR})$ decreased continuously. When H is oriented fairly close to h_{rf} (typically less than 5°) one observes the LR clearly resolved, however superposed on the small residual absorption of the UR for the reasons indicated previously.

The other results obtained are the following.

(1) The number of samples investigated was much smaller at 17 and 35 GHz than at 9.8 GHz. The overall data obtained presently are in favor of the assumption that $H_{\perp}^{\text{LR}}$ and $\Delta H_{\perp}^{\text{LR}}$, while they vary strongly with the working frequency, are independent up to the first order of the concentration of the Tb.

(2) The variation representative of the resonance-field $H_{\perp}^{\text{LR}}$ as a function of the working frequency f is reported in Fig. 3: $H_{\perp}^{\text{LR}}$ increases largely with increasing f, as expected theoretically,[1–4] but the experimental relationship differs from the theoretically computed ones, for the reasons discussed hereafter.

(3) An important experimental outcome, because it shows the exact nature of the LR, is the variation of N as a function of f. N decreases very rapidly with f, so the corresponding $H_{\perp}^{\text{LR}}$ increases. Actually we cannot establish a relationship having a general validity so the effect is just illustrated for a particular sample in Fig. 3.

Considering the experimental data, the following model can be proposed: In a ferromagnet with weak random anisotropy, a resonance mode specific to this magnetic state can be observed if the sample exhibits a very well defined H_k. H_k has several functions as regards to the possibility to detect the mode and the nature of the mode.

(4) The calculations about the LR were founded implicitly upon the hypothesis that it can be exited in a basically well-aligned ferromagnetic system where the random anisotropy can be treated as a perturbation.[1,2] This is the ferromagnet with wandering axis (FWA) regime.[3] Following a first evaluation[3] it was estimated that a FWA regime in a FWRA should be attained for a relatively small applied field. However, a later computation by Saslow showed[2] that the establishment of a well-aligned FWA regime is much more severe than originally thought, so the magnitude of the field H_a necessary to reach this regime must be much higher. Obviously, if H^{LR} is lower than H_a the detection of the LR could be very difficult or even impossible, because the presence of large magnetic inhomogeneities which contribute to the relaxation process. These problems are solved in a straightforward way by the existence of H_k. As shown by TBIS measurements and domain structure studies, we have a well-aligned system even when the magnetic field is reduced to

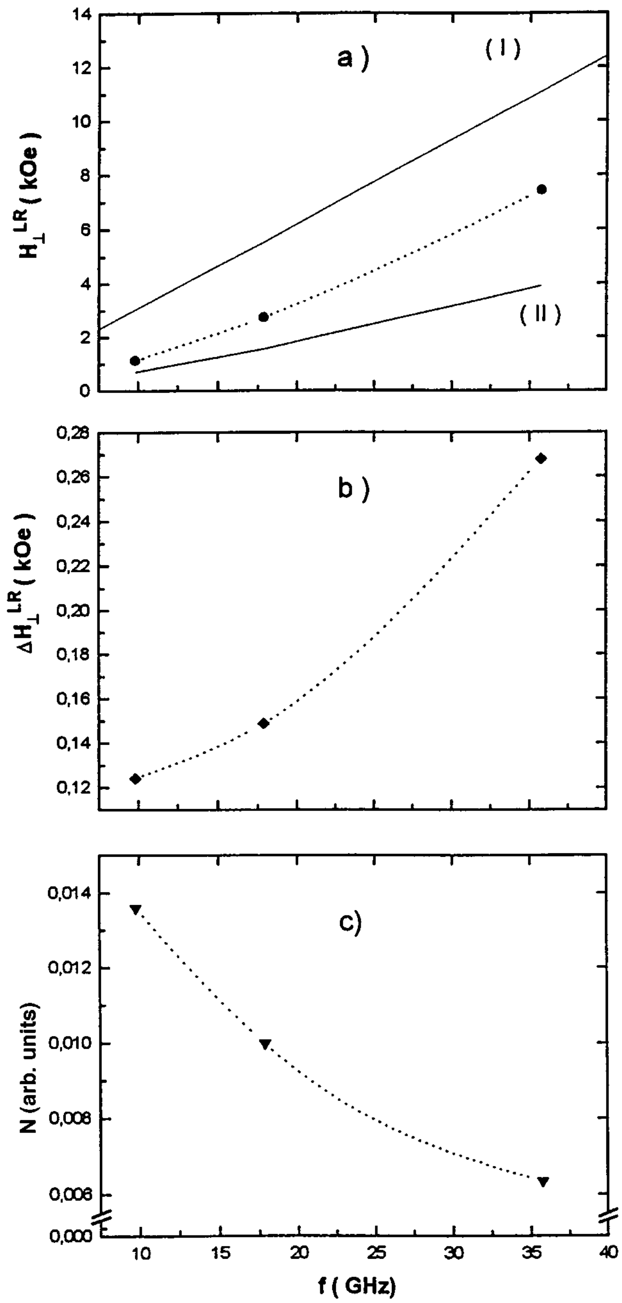

FIG. 3. The frequency dependence of (a) the resonance field (H^{LR}), (b) linewidth (ΔH^{LR}), and (c) microwave absorption ratio $N = I^{\text{LR}}(\Delta H^{\text{LR}})^2/I^{\text{UR}}(\Delta H^{\text{UR}})^2$. The dashed lines joining the experimental data are guides for eyes. (I) and (II) are computed curves with $H = \omega/\gamma$ and $H = (1/H_{ex})^{1/3}(\omega/\gamma)^{4/3}$ [see Eqs. (4) and (6) of Ref. 4]. $H_{ex} = 2.5 \times 10^5$ Oe (Ref. 5).

zero. Effectively at $H=0$ domain splitting with a configuration typical of a film with an in-plane easy axis is observed showing that the system is in a FWA regime. The random anisotropy now can be definitely considered as a perturbation and one of the main requirements necessary for the detection of LR is satisfied.

(5) The most logical way to explain N and its variations with f is that it corresponds to the ratio of the amount of spins contributing to the LR and UR, respectively. Whatever f, the UR is a collective process in which all spins participate. Consequently, the LR is a localized mode. The localized behavior of a resonance mode related to the random anisotropy is not a surprising phenomenon. Wave localization is just a general consequence of the disorder and is a special case of the localization process proposed originally by Anderson. The localization of spin waves were predicted in several computations, for various experimental situations,[4,8,9] and the present experiment is just the one where such a mode is observed directly.

A better understanding of the localization mechanism can be obtained by taking into account the particular magnetic structure of our films. Thin films which exhibit a local K_l and a uniaxial K_u anisotropy are formed of magnetically fairly strongly coupled[10] and geometrically very well-defined[10,11] regions. According to computations developed for crystalline films[10,11] and extended to amorphous ones,[12] these regions are an elongated rhombus[11] with the short axis parallel and the long axis perpendicular to the mean direction of the magnetization. For the parameters of our films the transverse axis is an order of magnitude longer than the longitudinal one. A current hypothesis is that the wavelength of the localized mode is related to the ferromagnetic coupling length. Presently it should correspond to the wavelength of the short-range ripple[10] given by $\lambda \sim 2\pi R_{LR} \sim 2\pi\sqrt{A}/[K_u h(\alpha')]$ (A, exchange const.). If h is applied along the easy axis $\alpha'=0$ and $h(0)=(H/H_k)+1$, so λ decreases when H is higher. General considerations on localization show that it changes gradually towards a ''stronger'' one as the localization length is shorter.[13] This mechanism allows one to understand that the amount of spins participating in the LR, so N decreases when the field required for resonance is higher. This localized nature of the LR is probably at the origin of the difference between the computed and the experimental value of H^{LR}: The computation of H^{LR} for various magnetic regimes[1,2,4] have been performed by assuming that the LR is a collective process.

(6) A very important property of H_k is that the nature of the mode is determined by the orientation of M_s with respect to H_k. Accordingly, the longitudinal nature of the mode is unchanged whatever the direction of M_s in the plane formed by $\vec{n}$ and H_k. The constant value of H^{LR} could be due to the localized nature of the mode: Only a very limited amount of spins contribute to the LR, hence the related demagnetizing field is negligible. On the other hand, the nature of the mode changes completely when H is rotated in the plane perpendicular to that formed by $\vec{n}$ and $\mathbf{H_k}$ as will be shown elsewhere.[14]

In conclusion, in a certain class of amorphous thin films which exhibit an in-plane uniaxial anisotropy field, a resonance mode related to the random anisotropy is detected. The mode displays the properties of a longitudinal resonance when the dc field is applied in the plane formed by $\vec{n}$ and $\mathbf{H_k}$ so observed experimentally when H is applied parallel to h_{rf}. The mode is localized and evolves towards ''stronger'' localization as the field required for resonance increases.

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